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Modified Hydrocarbon Resin and Use Thereof

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(54) [Title of the Invention] **Modified Hydrocarbon Resin and Use Thereof**

(57) [Summary]

[Object] An object of the present invention is to further improve compatibility with acrylic resins and other polar resins and to improve adhesive strength of adhesives by means of using a specific modified hydrocarbon resin as a tackifier.

[Means of Achievement] A modified hydrocarbon resin having a weight-average molecular weight of 300 to 3,000, obtained by means of the copolymerization of a vinyl aromatic hydrocarbon and a fraction that contains unsaturated hydrocarbons with 4 or 5 carbons and that is a byproduct of petroleum distillation, petroleum cracking, and the like in the presence of a Friedel-Crafts catalyst, wherein this hydrocarbon resin is obtained by means of the graft copolymerization of 0.1 to 10 wt% of an unsaturated carboxylic acid ester with a hydrocarbon resin that comprises 2 to 100 parts by weight of structural units based on unsaturated

hydrocarbons with 4 or 5 carbons, per 100 parts by weight of structural units based on the vinyl aromatic hydrocarbon; and a tackifier for acrylic adhesives comprising this resin.

[Claims]

[Claim 1] A modified hydrocarbon resin having a weight-average molecular weight of 300 to 3,000, obtained by means of the copolymerization of a vinyl aromatic hydrocarbon and a fraction that contains unsaturated hydrocarbons with 4 or 5 carbons and that is a byproduct of petroleum distillation, petroleum cracking, and the like in the presence of a Friedel-Crafts catalyst, wherein said hydrocarbon resin is obtained by means of the graft copolymerization of 0.1 to 10 wt% of an unsaturated carboxylic acid ester with a hydrocarbon resin that comprises 2 to 100 parts by weight of structural units based on unsaturated hydrocarbons with 4 or 5 carbons, per 100 parts by weight of structural units based on the vinyl aromatic hydrocarbon.

[Claim 2] The modified hydrocarbon resin according to claim 1, characterized in that the vinyl aromatic hydrocarbon is isopropenyl toluene.

[Claim 3] The modified hydrocarbon resin according to claim 1, characterized in that the unsaturated carboxylic acid alkyl ester is a dialkyl ester of an unsaturated dicarboxylic acid.

[Claim 4] The modified hydrocarbon resin according to claim 1, characterized in that the unsaturated carboxylic acid alkyl ester is di-n-dibutyl fumarate.

[Claim 5] The modified hydrocarbon resin according to claim 1, characterized in that the unsaturated carboxylic acid alkyl ester contains hydroxyl groups.

[Claim 6] The modified hydrocarbon resin according to claim 5, characterized in that the unsaturated carboxylic acid alkyl ester containing hydroxyl groups is 2-hydroxypropyl acrylate or 2-hydroxyethyl acrylate.

[Claim 7] A tackifier for acrylic adhesives comprising a modified hydrocarbon resin having a weight-average molecular weight of 300 to 3,000, obtained by means of the copolymerization of a vinyl aromatic hydrocarbon and a fraction that contains unsaturated hydrocarbons with 4 or 5 carbons and that is a byproduct of petroleum distillation, petroleum cracking, and the like in the presence of a Friedel-Crafts catalyst, wherein said hydrocarbon resin is obtained by means of the

graft copolymerization of 0.1 to 10 wt% of an unsaturated carboxylic acid ester with a hydrocarbon resin that comprises 2 to 100 parts by weight of structural units based on unsaturated hydrocarbons with 4 or 5 carbons, per 100 parts by weight of structural units based on the vinyl aromatic hydrocarbon.

[Detailed Description of the Invention]

[0001]

[Technological Field of the Invention] The present invention relates to a novel modified hydrocarbon resin and a tackifier for acrylic pressure-sensitive adhesives composed thereof.

[0002]

[Prior Art] Applications that involve adhesives can be cited as typical applications in which tackifying resins are used. Low-molecular-weight polymers (such as styrene, vinyl toluene, α -methyl styrene, aliphatic petroleum resins, aromatic petroleum resins, rosin, modified rosin, terpene resins, coumarone resins, phenol resins, terpene-phenol resins, coumarone-indene resins, and xylene resins) are being studied for use as the tackifying resins of acrylic pressure-sensitive adhesives. Nevertheless, the above-mentioned adhesives have a disadvantage when used as acrylic pressure-sensitive adhesives in that sufficient adhesive strength and bonding strength are not realized because compatibility is not satisfactory. Moreover, petroleum resins, rosin, terpene-phenol resin, coumarone-indene resin, and other materials produced from ordinary unsaturated hydrocarbon fractions have problems in terms of color, thermal stability, and the like.

[0003] The inventors have previously proposed a product obtained by means of polymerizing isopropenyl toluene in a specific ratio with a fraction that has 4 or 5 carbons and is a byproduct of petroleum distillation or petroleum cracking, and also proposed that an isopropenyl toluene homopolymer be used in order to improve on these problems (JP (Kokai) 49-118729, JP (Kokai) 49-128945, JP (Kokoku) 54-34033). These hydrocarbon resins have an excellent initial hue, heat-resistant hue, and thermal stability, and they also have better compatibility with acrylic resins than do low-molecular-weight polymers such as vinyl toluene, α -methyl styrene, and petroleum resins. Therefore, these hydrocarbon resins are used as tackifiers for acrylic pressure-sensitive adhesives.

[0004]

[Problems to Be Solved by the Invention] Nevertheless, market demands in recent years have pointed to the insufficient adhesive strength of adhesives. Therefore, an object of the present invention is to further improve compatibility with acrylic resins and other polar resins, and to enhance the adhesive strength of adhesives by using a specific modified hydrocarbon resin as a hydrocarbon resin that is also used as a tackifier.

[0005]

[Means Used to Solve the Above-Mentioned Problems] In order to solve the above-mentioned problems, the present invention provides a modified hydrocarbon resin that has a weight-average molecular weight of 300 to 3,000 and is obtained by means of the copolymerization of a vinyl aromatic hydrocarbon and a fraction that contains unsaturated hydrocarbons with 4 or 5 carbons and that is a byproduct of petroleum distillation, petroleum cracking, and the like in the presence of a Friedel-Crafts catalyst, wherein this hydrocarbon resin is obtained by means of the graft copolymerization of 0.1 to 10 wt% of an unsaturated carboxylic acid ester with a hydrocarbon resin that comprises 2 to 100 parts by weight of structural units based on unsaturated hydrocarbons with 4 or 5 carbons per 100 parts by weight of structural units based on the vinyl aromatic hydrocarbon; and a tackifier for acrylic adhesives comprising this resin.

[0006] Styrene, α -methyl styrene, vinyl toluene, isopropenyl toluene, or the like can be cited as the vinyl aromatic compound that is used in the present invention, but a hydrocarbon resin with a particularly narrow molecular weight distribution and particularly good properties can be obtained when isopropenyl toluene is used. An ortho, meta, or para isomer or and a mixture of these is normally used as the isopropenyl toluene comprising an isopropenyl toluene polymer, but it is preferred that a mixture of 20 to 60 wt% of the para isomer, 40 to 80 wt% of the meta isomer, and 0 to 10 wt% of the ortho isomer be used.

[0007] Any fraction that has unsaturated hydrocarbons with 4 or 5 carbons and is a byproduct of petroleum distillation or petroleum cracking can be used as the unsaturated hydrocarbon with 4 or 5 carbons that is employed in the present invention. Fractions containing hydrocarbons that have 4 or 5 carbons (C4 and C5 fractions hereafter) and are a byproduct of petroleum distillation or petroleum cracking have boiling points that usually range under normal pressure from -15 to

+45°C, and these include polymeric monomers such as 1-butene, isobutene, 2-butene, 1,3-butadiene, 1-pentene, 2-methyl-1-butene, 3-methyl-1-butene, 2-pentene, isoprene, 1,3-pentadiene, cyclopentadiene, and the like. In the present invention, it is possible to use any fraction that contains polymeric monomers selected from C4 and C5 fractions, that is, the C4 and C5 fractions, the C4 fraction, a C4 fraction from which butadiene has been removed, the C5 fraction, a C5 fraction from which isoprene has been removed, a C5 fraction from which cyclopentadiene has been removed, and the like. The hydrocarbon resin that is used in the present invention is obtained as described above by means of the copolymerization of 2 to 100 parts by weight, preferably 5 to 70 parts by weight, of an unsaturated hydrocarbon with 4 or 5 carbons per 100 parts by weight of a vinyl aromatic compound.

[0008] The copolymerization reaction between the two constituent components is preferably performed in the presence of a Friedel-Crafts catalyst. Conventional Friedel-Crafts catalysts can be used. Specific examples include aluminum chloride, aluminum bromide, dichloromonoethyl aluminum, titanium tetrachloride, tin tetrachloride, boron trifluoride, boron trifluoride, various other complexes, and the like. The amount in which the Friedel-Crafts catalyst is used is usually within a range of 0.05 to 5 parts by weight, preferably 0.1 to 2 parts by weight, per a total of 100 parts by weight of starting material.

[0009] The copolymerization reaction between the two structural components should be performed using a solvent so that the concentration of the polymeric monomer is brought to 10 to 60 wt% in order to eliminate the heat of the reaction, to control the liquid viscosity of the reaction, and to adjust the molecular weight. Examples of appropriate solvents include aliphatic hydrocarbons such as pentane, hexane, heptane, and octane; alicyclic hydrocarbons such as cyclopentane, cyclohexane, and methyl cyclohexane; aromatic hydrocarbons such as toluene, xylene, ethyl benzene, and mesitylene; and mixtures thereof. The polymerization temperature varies with the starting material composition but is preferably within a range of -50 to +50°C. Moreover, it is preferred that the reaction time is usually within a range of 10 minutes to 10 hours. Once polymerization is over, the catalyst is decomposed with an aqueous alkali solution, methanol, and the like, the product is rinsed with water, and the unreacted starting material, solvent, and the like are then removed by means of stripping in order to obtain the desired hydrocarbon resin. The softening point of the resin that is obtained in this way is usually

30 to 150°C (ring and ball method). Moreover, the molecular weight is usually within a range of 300 to 5,000 (in terms of weight-average molecular weight).

[0010] At least one type selected from unsaturated carboxylic acid alkyl esters is used as the modifying monomer that is graft polymerized with the above-mentioned hydrocarbon resin in order to obtain the modified hydrocarbon resin of the present invention. An unsaturated carboxylic acid alkyl ester is used as the monomer for such graft copolymerization. Specific examples include alkyl esters of unsaturated monocarboxylic acids, such as acrylic acid and methacrylic acid; and alkyl esters of unsaturated dicarboxylic acids, such as maleic acid, fumaric acid, itaconic acid, crotonic acid, isocrotonic acid, citraconic acid, allyl succinic acid, mesaconic acid, glutaconic acid, nadic acid (endocis-bicyclo[2,2,1]hepto-2-ene-5,6-dicarboxylic acid), methylnadic acid, tetrahydrophthalic acid, and methylhexahydrophthalic acid. In addition to alkyls with 1 to 4 carbons, such as methyl, ethyl, n-propyl, i-propyl, n-butyl, i-butyl, sec-butyl, t-butyl, and the like, alkyls where hydroxyl groups, halogens, and the like are substituted for the alkyl groups can be cited as the alkyls of the alkyl ester.

[0011] Hydroxyethyl (meth)acrylate, 2-hydroxypropyl (meth)acrylate, 3-hydroxypropyl (meth)acrylate, 2-hydroxy-3-phenoxypropyl (meth)acrylate, 3-chloro-2-hydroxypropyl (meth)acrylate, glycerin mono(meth)acrylate, pentaerythritol mono(meth)acrylate, trimethylolpropane mono(meth)acrylate, tetraethylolethane mono(meth)acrylate, butanediol mono(meth)acrylate, polyethylene glycol mono(meth)acrylate, and the like can be cited as specific examples of the unsaturated carboxylic acid alkyl ester comprising hydroxyl groups in the ester moiety.

[0012] Of the above-mentioned monomers, di-n-butyl fumarate, hydroxyethyl (meth)acrylate, and 2-hydroxypropyl acrylate are preferred. These monomers can be used alone, or two or more types can be simultaneously used. Moreover, another vinyl monomer, such as styrene, α -methyl styrene (meth)acrylonitrile, (meth)acrylamide, vinyl acetate, or the like, can be co-grafted with these monomers. The amount grafted is preferably 0.1 to 10 wt%, and particularly preferably 1 to 8 wt%, of the hydrocarbon resin. If the amount grafted is less than 0.1 wt%, the result will be lower compatibility with the acrylic copolymer or another polar polymer as the base polymer of an adhesive. Moreover, when the amount grafted is more than 10 wt%, there may be an

improving effect on compatibility, but there will also be a considerable reduction in the softening point.

[0013] Various methods can be cited as examples of methods for the graft copolymerization of at least one type of modifying monomer selected from unsaturated carboxylic acid alkyl esters with the above-mentioned hydrocarbon resin. These examples include methods whereby a hydrocarbon resin is dissolved in an organic solvent, the above-mentioned modifying monomer and radical polymerization initiator are added in succession, and the product is graft copolymerized by means of heating and stirring; methods whereby this hydrocarbon resin is heated and melted, a modifying monomer and a radical polymerization initiator are added to the resulting melt, and the product is graft copolymerized by means of stirring; methods whereby a hydrocarbon resin, modifying monomer, and radical initiator are pre-mixed, the resulting mixture is fed to an extruder, and graft polymerization is performed as the mixture is being heated and kneaded; and methods whereby after this hydrocarbon resin in tablet form has been impregnated by a solution that is made by means of dissolving a modifying monomer and radical polymerization initiator in an organic solvent, graft copolymerization is performed by heating the hydrocarbon resin in tablet form to the highest temperature at which it will not dissolve. In addition, if unsaturated bonds of this hydrocarbon resin are still present, the radical polymerization initiator may not be necessary and the above-mentioned graft copolymerization may be performed in the absence of a radical polymerization initiator. The ideal reaction temperature is 50°C or higher, particularly 80 to 200°C, and the reaction time is 2 to 10 hours. The reaction system can be either a batch system or a continuous system, but the batch system is preferred for uniform graft copolymerization.

[0014] Any radical polymerization initiator can be used for graft copolymerization using a radical polymerization initiator as long as it promotes the reaction between the block copolymer and the above-mentioned graft copolymer unit, but organic peroxides and organic peresters are particularly preferred. Specific examples include benzoyl peroxide, dichlorobenzoyl peroxide, dicumyl peroxide, di-tert-butyl peroxide, 2,5-dimethyl-2,5-di(peroxyphenyl)benzene, hexyl-3,1,4-bis(tert-butylperoxyisopropyl)benzene, lauroyl peroxide, tert-butyl peracetate, 2,5-dimethyl-2,5-di(tert-butylperoxy)hexene-3,2,5-dimethyl-2,5-di(tert-butyl peroxide)hexane, tert-butylbenzoate, tert-butylperphenyl acetate, tert-butyl perisobutyrate, tert-butylper-sec-octoate, tert-

butyl perpivalate, cumyl perpivalate, tert-butyl perdiethylacetate, as well as azo compounds such as azobisisobutylnitrile and dimethyl azobisisobutyronitrile. Preferred among these are dialkyl peroxides, including dicumyl peroxide, di-tert-butyl peroxide, 2,5-dimethyl-2,5-di(tert-butylperoxy)hexene-3,2,5-dimethyl-2,5-di(tert-butylperoxy)hexane, and 1,4-bis(tert-butylperoxyisopropyl)benzene. The amount of this type of radical polymerization initiator is usually 0.1 to 5 wt% in relation to the starting hydrocarbon resin.

[0015] The weight-average molecular weight of the modified hydrocarbon resin obtained in this way is 300 to 3,000, preferably 500 to 2,500. If the molecular weight is less than 300, the modified product will have pronounced blocking properties and will be difficult to handle. Moreover, if the molecular weight is greater than 3,000, the melt viscosity of the modified hydrocarbon resin will be high and work performance will be poor. Furthermore, the tackiness of the tackifying resin will decline considerably. The hue of this modified hydrocarbon resin should be 3 or lower, particularly 2 or lower, as determined by the Gardner hue determination method. Conventional compounding agents, such as softeners, stabilizers, fillers, antioxidants, and the like, can also be added to the modified hydrocarbon resin of the present invention as long as the addition is within a range that does not compromise the object of the present invention.

[0016] The modified hydrocarbon resin of the present invention has excellent compatibility with polar resins and can therefore be used as a tackifier for acrylic adhesives. There are no special restrictions to the acrylic polymer that is the primary component of the acrylic adhesive, and various homopolymers and copolymers known as this type of pressure-sensitive adhesive composition in the past can be used as is. Various (meth)acrylic acid esters can be cited as such monomers. Methyl (meth)acrylate, ethyl (meth)acrylate, butyl (meth)acrylate, 2-ethylhexyl (meth)acrylate, and the like can be specifically cited, and these can be used alone or in combination with one another. Moreover, it is also possible to copolymerize a copolymerizable monomer, specifically vinyl acetate, styrene, and the like, as needed. Furthermore, glycidyl (meth)acrylate and N-methylol(meth)acrylamide may be jointly used to provide crosslinking reactivity. When an acrylic polymer with crosslinking reactivity is used, polyamine, polyisocyanate, melamine resin, urea resin, epoxy resin, and the like may be used as needed for the crosslinking agent.

[0017] There are no special restrictions to the method of producing the above-mentioned acrylic polymer, and various conventional methods are used. For instance, polymerization can be easily performed by means of a radical polymerization, such as bulk polymerization, solution polymerization, or suspension polymerization. The above-mentioned acrylic polymer and modified hydrocarbon resin of the present invention are usually combined in a ratio of 95:5 to 50:50 in terms of solid content. The acrylic adhesive obtained in this way will have excellent hue and improved adhesive strength. Thus, labels, adhesive tape, adhesive double coated tape, and other products obtained using the acrylic adhesive obtained in this way can be employed for a wide range of adherends, including polyolefins. Moreover, the applications of this modified hydrocarbon resin are not limited to those cited and may also include, for instance, hot melt adhesives, paints, inks, sizing agents for paper, rubber softeners, and the like.

[0018] Working examples of the present invention are given below.

<Synthesis of Hydrocarbon Resin>

(Reference Example 1) One hundred grams of isopropenyl toluene (purity of 99.2%) obtained by means of the distillation and separation from the acidolysis of cymene hydroperoxide, 10 g of a fraction containing C4 and C5 unsaturated hydrocarbons obtained by means of the pyrolysis of petroleum naphtha, and 150 g of toluene were introduced into an autoclave during the cymene cresol process, and 0.3 g of BF₃ phenol complex was added dropwise as the catalyst over a period of approximately 10 minutes while the system was stirred and kept at a temperature of 0°C. Stirring was continued for another three hours. Next, 50 mL of an aqueous 5 wt% sodium hydroxide solution were added and vigorously stirred for 30 minutes to decompose the catalyst. The aqueous phase was separated and the product was rinsed with water until the polymerization oil became neutral. Then the unreacted oil and solvent toluene were distilled off while kept at a reduced pressure and heated to obtain a white mass of hydrocarbon resin. The composition of the starting monomer is as shown below. Moreover, the properties of the resulting hydrocarbon resin are shown in Table 1.

[0019]

<u>Isopropenyl toluene composition</u>	<u>wt%</u>
Ortho-isopropenyl toluene	5.3
Meta-isopropenyl toluene	60.4
Para-isopropenyl toluene	33.5
Cymene	0.8

C4, C5 fraction composition

C3	1.5
Isobutene	0.4
n-Butane	1.3
1-Butene	10.5
Isobutylene	19.8
Trans-2-butene	8.7
Cis-2-butene	4.4
1,3-Butadiene	20.9
n-Pentane	5.1
Isoprene	5.6
1,3-Pentadiene	2.8
Cyclopentadiene	3.1
Others (fractions with boiling points of 80°C or lower)	15.9

[0020] (Reference Example 2) Except that the amount of BF₃ phenol complex added in Reference Example 1 was brought to 0.25 g, a hydrocarbon resin was synthesized as in Reference Example 1. The properties of the resulting hydrocarbon resin are shown in Table 1. The resin properties were determined by means of the following methods.

Hue:	Gardner	ASTM D 154-58
Softening point:	Ring and ball method	
Molecular weight:	GPC (in terms of polystyrene)	

[0021]

[Table 1]

	Hue	Softening point	Weight-average molecular weight	Bromine number
Reference Example 1	1	96	1310	15
Reference Example 2	1	110	1630	10

[0022] <Preparation of Acrylic Polymer>

(Reference Example 3: Butyl acrylic polymer)

One hundred grams of ethyl acetate were introduced into a reactor with a stirring device, cooling tube, funnel, and nitrogen tube, and the temperature was raised to 80°C. Then, a mixture comprising 97 g of butyl acrylate, 3 g of acrylic acid, and 0.2 g of benzoyl peroxide was loaded in advance into the funnel and added dropwise from the funnel over a period of three hours in a nitrogen stream. The product was further heated and stirred for one hour while the established temperature was maintained in order to complete the reaction. The solid concentration of the resulting polymer was 50%.

(Reference Example 4: 2-Ethyl hexyl acrylic polymer) Benzoyl R-50 (solid concentration of 44 wt%) made by Yushi Kogyo Co., Ltd. was used.

[0023]

<Working Example 1>

First, 250 g of hydrocarbon resin that had been synthesized in Reference Example 1 were introduced into a reactor with a stirring device, funnel, and nitrogen tube, and the inside of the system was purged with nitrogen for 15 minutes. The device, while still filled with nitrogen, was heated to 170°C, and the hydrocarbon resin was melted. Next, 1.3 g of ditertiary butyl peroxide (DTBPO hereafter) and 13 g of di-n-butyl fumarate (di-butyl fumarate hereafter) were added dropwise over a period of two hours, and once dropwise addition had been completed, the contents were reacted for another hour to complete the reaction. When the reaction was over, the unreacted dibutyl fumarate and DTBP decomposition product were removed by being kept under reduced pressure for 30 minutes under conditions of 180°C and 10 mmHg. The extent of dibutyl fumarate modification of the resulting modified hydrocarbon was 3.9 wt% based on oxygen

analysis. The hue was 1. The reaction conditions and resin properties are shown in Tables 2 and 3.

[0024]

<Working Examples 2 through 7>

A graft reaction was conducted under the same conditions as in Working Example 1 using the same equipment as in Working Example 1, with the hydrocarbon resin and graft monomer being varied. The reaction conditions and properties of the resulting modified hydrocarbon resin are shown in Tables 2 and 3.

[0025]

[Table 2]

	Starting material	Graft monomer	Amount (g)	Peroxide	Amount (g)
Working Example 1	Reference Example 1	Dibutyl fumarate	13.2	DTBPO	1.3
Working Example 2	Reference Example 1	Dibutyl fumarate	27.7	↑	2.77
Working Example 3	Reference Example 2	Dibutyl fumarate	13.2	↑	1.3
Working Example 4	Reference Example 2	Dibutyl fumarate	27.7	↑	2.77
Working Example 5	Reference Example 1	2-Hydroxypropyl acrylate	2.5	↑	0.25
Working Example 6	Reference Example 1	2-Hydroxypropyl acrylate	12.5	↑	1.25
Working Example 7	Reference Example 1	2-Hydroxyethyl acrylate	12.5	↑	1.25

[0026]

[Table 3]

	Extent of modification (wt%)	Softening point (°C)	Weight-average molecular weight	Hue
Working Example 1	3.5	78	1230	1↓
Working Example 2	6.9	67	1250	1.5
Working Example 3	3.6	96	1350	1↓
Working Example 4	7.0	87	1400	1↓
Working Example 5	0.6	95	1200	1↓
Working Example 6	4.2	94	1300	1↓
Working Example 7	4.1	87	1300	1↓

[0027] <Preparation of acrylic adhesive and evaluation of adhesive strength>

Evaluation was performed as follows.

Appearance of adhesive: Ranked in three steps of O, Δ, and X by macroscopic evaluation.

Adhesive strength: Evaluated by 180° peeling tests.

<Working Example 8> Once the acrylic polymer synthesized in Reference Example 3 and the modified hydrocarbon polymer synthesis in Working Example 1 were mixed in a ratio of 90/10 by solid content and thoroughly stirred, 2.7 parts of Coronet L (polyisocyanate compound: Nippon Polyurethane Co., Ltd.) were added to make an acrylic adhesive. The adhesive had a transparent appearance. This adhesive was applied to a 40-μ-thick PET film in a dry thickness of 30 μ and dried for five minutes at 80°C to make an adhesive tape. This adhesive tape was applied to a PET film under a pressure of 500 g to obtain a sample, and adhesive strength was measured. The results are shown in Table 4.

<Working Examples 9 through 12>

Except that the combination of modified hydrocarbon resin and acrylic polymer was varied, samples were made and adhesive strength was determined by means of the same method as in Working Example 8. The results are shown in Table 4.

[0028] <Comparative Example 1> Except that the modified hydrocarbon resin was not used, a sample was prepared and adhesive strength determined by means of the same method as in Working Example 8. The results are shown in Table 4.

<Comparative Examples 2 and 3> Except that the hydrocarbon resins in Reference Examples 1 and 2 were used in place of the modified hydrocarbon resin, samples were prepared and adhesive strength was determined by means of the same method as in Working Example 8. The results are shown in Table 4.

[0029]

[Table 4]

	Modified hydrocarbon resin	Acrylic polymer	Appearance	Adhesive strength (g/25 mm)
Working Example 8	Working Example 1	Reference Example 3	○	1600
Working Example 9	Working Example 3	Reference Example 3	○	1880
Working Example 10	Working Example 3	Reference Example 4	△	1720
Working Example 11	Working Example 6	Reference Example 3	○	1650
Working Example 12	Working Example 6	Reference Example 4	△	1500
Comparative Example 1	—	Reference Example 3	—	1010
Comparative Example 2	Reference Example 1	Reference Example 3	△	1280
Comparative Example 3	Reference Example 2	Reference Example 3	△	1350